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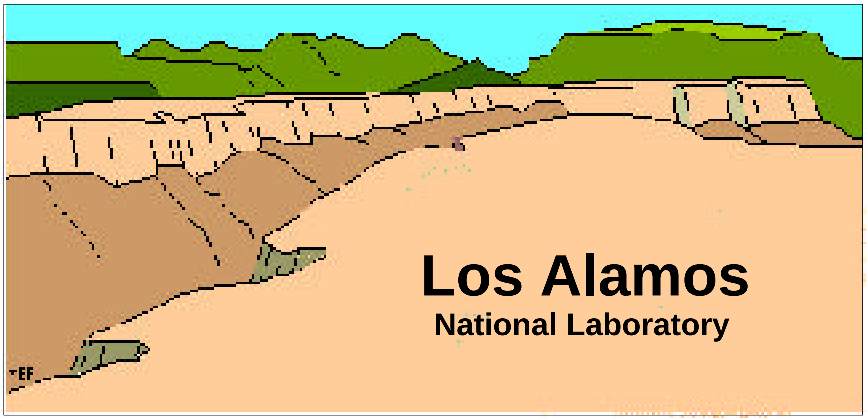

Analysis of SX Farm Leak Histories
—Historical Leak Model (HLM)

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Summary:
This report uses readily available historical information to better define the volume, chemical composition, and Cs-137/Sr-90 amounts for leaks that have occurred in the past for tanks SX-108, SX-109, SX-111, and SX-112. In particular a Historical Leak Model (HLM) is developed that is a month by month reconciliation of tank levels, fill records, and calculated boil-off rates for these tanks. The HLM analysis is an independent leak estimate that reconstructs the tank thermal histories thereby deriving each tank's evaporative volume loss and by difference, its unaccounted losses as well. The HLM analysis was meant to demonstrate the viability of its approach, not necessarily to establish the HLM leak estimates as being definitive. Past leak estimates for these tanks have invariably resorted to soil wetting arguments but the extent of soil contaminated by each leak has always been highly uncertain. There is also a great deal of uncertainty with the HLM that was not quantified in this report, but will be addressed later.

These four tanks (among others) were used from 1956 to 1975 for storage of high-level waste from the Redox process at Hanford. During their operation, tank waste temperatures were often as high as 150°C (300°F), but were more typically around 130°C. The primary tank cooling was by evaporation of tank waste and therefore periodic replacement of lost volume with water was necessary to maintain each tank's inventory. This active "reflux" of waste resulted in very substantial turnovers in tank inventory as well as significant structural degradation of these tanks. As a result of the loss of structural integrity, each of these tanks leaked during their active periods of operation. Unfortunately, the large turnover in tank volume associated with their reflux cooling has made a determination of leak volumes very difficult.

During much of these tanks' operational histories, inventory losses because of evaporative cooling could have effectively masked any volume loss due to leak. However, careful comparison with reported tank levels during certain periods clearly show unaccounted volume losses for many tanks. As a result of the HLM analysis, SX-108, SX-109, SX-111, and SX-112 all show clear evidence of unaccounted volume losses during the period 1958 to 1975. Likewise, the HLM does not show similar unaccounted volume losses for tank SX-105, a tank with no reported leak history, verifying that the HLM is consistent with SX-105 not leaking.

These unaccounted volume losses establish the leak start date and rate, and when propagated over time show that SX-108 lost 203 kgal followed by SX-109 at 111, SX-111 at 55, and SX-112 at 44 kgal. These leak volumes represent maximum or upper bounds estimates of each leak and are in total volume about six times the previous leak estimates. Minimum leak estimates are about 50% of these values based on judgments about the heat and leak rate uncertainties. Except for tank SX-112, these results all assume that leaks are continuous from start to end and depend only on three parameters: an elevation in kgal, a rate in kgal per month per kgal above that elevation, and a start date for the leak.

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Acknowledgments:
This work has been performed under the auspices of the Department of Energy under contract to the University of California.

Scope:
To use best information currently (as of October 1996) available to estimate the leak volumes for four SX Farm tanks, SX-108, SX-109, SX-111, and SX-112. Once leak volumes and dates have been established, derive curies of Cs-137 and Sr-90 as well as leak chemical composition for major analytes (Na^+, OH^-, CrO_4^{2-}, NO_3^-, NO_2^-, AlO_2^-).

One must recognize that there are multiple unstated assumptions and uncertainties with all existing leak estimates. The HLM is simply an attempt at deriving an independent estimate for a particular set of waste tanks, boiling waste tanks. The HLM analysis was meant to demonstrate the viability of this approach, not necessarily to establish the HLM leak estimates as being definitive.

I. Background:
Single-Shell Tanks (SST's) that have leaked various amounts of high level waste into the soil column have been a fairly common past occurrence at the Hanford Site. All told, some 67 of the 149 SST's have reportedly leaked (45%). These waste tanks at Hanford had been used for a variety of waste storage missions and of all of these missions, none were more stressful to the tanks than that of containing boiling or aging wastes. The combination of high temperatures, sudden steam "bumps", and caustic high nitrate, nitrite, and aluminate wastes all contributed to the degradation of these tanks.

The set of 25 tanks that contained aging waste (15 SX Farm, 6 A Farm and 4 AX Farm tanks) comprise a particular problem with respect to estimating leaks for two reasons. First, these tanks were cooled by evaporation of water as the waste boiled. Upwards of 200 kgal/mo of condensate was evaporated, cooled, and replaced for some of these tanks during their peak operations. Along with such large turnovers in volume, differentiating between a normal loss of water (i.e. makeup) in the process from that volume waste lost as leaks was extremely difficult. Of this set of 25 boiling waste tanks, 16 (64%) are reported leakers. Of the nine tanks in SX Farm that were heavily used for boiling waste, all have leaked.

As these wastes concentrated, precipitates of sodium nitrate, nitrite, and aluminate would produce cement-like scale on the cooler surfaces within each tank. Solid monoliths of salt have been reported in many tanks with concentrated waste. As a result, this scale would often seal leaks as the tank cooled and therefore further complicate any effective leak detection by material accountability, especially as the tank cooled.

As a result of these and other difficulties, all of the tank leaks in SX Farm were first detected in the soil and not from material accountability. That is, these leaks were only indicated after the fact as radioactive contamination appeared in vertical or lateral drywells around and under each tank. Thus, the period of time for peak heat load for each tank was the time it was most likely to leak and was exactly the most difficult period to deduce a leak by material lost. The HLM is an attempt to better define leak estimates for four SX Farm tanks by reconciling their fill histories with the volume evaporated by their heat loads.

II. Approach:
The Historical Leak Model (HLM) uses limited information for tank waste volumes and waste transactions that is now available on a quarterly basis (from 1956-60 on a monthly basis, from 1960-65 only semi-annually) over these tanks' histories (WSTRS). The HLM then reconciles this volume information with the evaporation rate that is calculated based on the total heat load of each tank. The historical tank heat loads are based on the radionuclides processed on a month by month basis (by Watrous and Wootan) with a calculation of the short-lived radionuclide heating based on the Cs-137 and Sr-90 fission products by use of effective decay heat curves for each batch processed.

Table 1 shows the radionuclide heating that is used. An effective cooling curve was derived by fitting an ORIGEN2 calculation of spent fuel heat generation decay and scaled relative to Cs-137 and Sr-90 fission products. We found that four surrogate radionuclides adequately represented the cooling rate within ±5% from 100 days to six years. After six years, the majority of the decay heat is due to Cs-137 and Sr-90 decay. This approximation allows us to recreate the heat generation rate of each load of fuel that was processed and decay those rates appropriately.

Table 1. Short-lived radionuclide heating.

radionuclide or surrogate	half-life (years)	heat production relative to Cs-137 or Sr-90
Cs-137	30.2	4.74e-3 W/Ci
Sr-90	28.5	6.70e-3 W/Ci
R1	1.0	12.9
R2	0.45	15.0
R3	0.22	108
R4	0.080	200

All of this information is placed into an Microsoft Excel workbook on a month-by-month basis and is termed the HLM spreadsheet. Within the HLM, account is not only made for short-lived radionuclides but also for tank-to-tank transactions, which can move significant amounts of radionuclide inventory from one tank to another. A further assumption is made about the soluble versus the insoluble fraction of radionuclide heating in order to allow moving fractions of the tank heat load among the tanks. All Cs-137 is assumed to reside in the supernatant and all Sr-90 is assumed to reside in the sludge. As a result of moving tank inventory, more tanks than just these four have been analyzed but are not included in this report.

Once unaccounted volume losses and dates for those losses are established, the HDW model provides estimates for leak compositions by assigning unaccounted volume losses to tank inventory leaked to ground. If more detailed transaction and volume information becomes available, the HLM leak estimates can be further refined.

The radionuclide data is expressed as activity in Ci decayed to 1-1-94 for each batch of fuel processed for the history of Hanford and each fuel batch includes a cooling time in days. We have taken that data and regrown the Cs-137 and Sr-90 to the waste addition date and binned the batches appropriately (monthly, quarterly, or semiannually) depending the transaction records reporting frequency. If there were waste additions to more than one tank in a given period, radionuclides were simply partitioned in proportion to the transaction volumes associated with each tank. This fraction is specified in the fr.rads. column in the spreadsheets in App. A.

Given the amount of Cs-137 and Sr-90 regrown for each batch of fuel processed, each of the four short-lived surrogates are calculated and correspondingly binned into the transaction period. Each surrogate radionuclide is decayed on a month-by-month basis in the spreadsheet according to its half-life. Liquid waste transfers from each tank as well as leaks to the soil column result in removal of only the soluble Cs-137 surrogate set while the insoluble Sr-90 surrogate set always remains in the tank into which it was first placed.

III. Methodology:

The SX Farm tanks were used to store high-level waste from the Redox Process (S Plant) with very large peak heat loads, upwards of 1-2 million Btu's/hr. These heat loads were largely a result of the short-lived radionuclides that were present in the fuel that was being processed. After approximately six years these early heat sources decayed leaving the 30-year isotopes Cs-137 and Sr-90, which represent only 0.5% of the heat load at 150 days cooling time. During the periods of extremely high heat load, tanks were cooled by reflux and large amounts of water boiled from the tank were returned as condensate. At peak heat loads, upwards of 200 kgal of waste inventory was evaporated per month of operation for some tanks and at peak times the rate amounted to some 450 kgal of condensate collectively for the entire tank farm. During some of these peak boil-off time periods, leaks on the order of 0.5 to 2.0 kgal per month appeared in many tanks and it would have been very difficult to discern these leaks as unaccounted material in the presence of such large volume turnovers from self boiling.

The Historical Leak Model attempts to reconcile reported tank volumes with those calculated based on expected volume reduction associated with historical heat loads for each tank's waste. The HLM calculates volume changes based on the evaporative cooling rate of a tank and reconciles those level changes with reported volumes and transactions for each tank during the same time period. Using this reconciliation, measured tank volumes that are greater than those expected because of evaporative loss are accounted. Measured tank volume losses less than those predicted by waste evaporation are assigned as unaccounted volume losses. These unaccounted volume losses may be due to normal volume loss, inaccuracies of the HLM, or waste inventory that has been lost to the soil through a breach in the tank liner.

For example, in January 1961, the evaporation rate for SX-108 was 2.0 kgal/mo and the unaccounted volume loss was 2.0 kgal/mo. This assumes a total tank heat of 38 kW, a heat loss to ground and air of 20 kW leaving an 18 kW evaporative loss. In principle, the leak rate could be as much as a factor of two lower, on the order of 1 kgal/mo, given a higher evaporation rate and a corresponding lower heat loss to ground. If the heat loss to ground were as low as 10 kW, the evaporation rate would be 50% greater (3.1 kgal/mo) lowering the leak rate to 0.9 kgal/mo. A critical factor is the tank waste temperature during this period. If tank waste temperature data show a significant decrease during this period, this will also lower the leak estimate. A decreased tank temperature suggests that evaporation is cooling that tank waste beyond what is necessary to remove the tank radionuclide heat load.

Tanks that were cooled by evaporation are assumed to have been on a reflux condenser system. In a total reflux, condensate derived from tank vapor is returned to each tank from a cooler. Although the details of this process are unclear at this time, generally the condensate was extracted from a number of tanks with a common ventilation system though separate isolated coolers were also available for individual tanks as well. In fact, specific mention is made for placing SX-107 and SX-108 on isolated coolers during periods of uncertainty about unaccounted volume losses for these two tanks. When tanks were on common ventilation, though, it was not possible to determine how much condensate actually came from each individual tank in the entire group.

In addition, there was inevitably some loss of water vapor out of the system during the process and this loss needed to be made up by periodic water additions back to the tanks. It is only when this added makeup volume exceeded some specified tolerance that any leak would be discernible from an accounted materials basis. Thus, a leak that was small on the order of the system throughput would be completely masked by the operation.

If a tank was allowed to self-concentrate, condensate would be directed to SX-106 instead of being returned to the tank. Often the condensate was partly refluxed and partly extracted, resulting in net concentration of a tank's waste inventory to some target volume. This condensate in SX-106 presumably was used for makeup additions. During this process, there was undoubtedly some loss of water vapor to the air and that loss needed to be made up by periodic additions of water back into each tank. This amount of makeup would likely scale with the total reflux rate of each tank. For example, a 2% loss would mean that SX-108 would need a 2.6 kgal/mo makeup in October 1963, since the reflux was evaporating 130 kgal/mo at that time. The unaccounted volume loss during that period was on the order of 1.5 kgal/mo and therefore the unaccounted volume was more than one-half of the entire makeup volume. The amount of unaccounted volume relative to the makeup volume is very important since it is the makeup volume that masks small tank leaks. Moreover, since the entire farm's evaporation rate was on the order of 200 kgal/mo, such a leak would have been effectively masked by the normal water makeup for the process.

The HLM assigns leak rates on the basis of unaccounted volume losses during periods of low reflux. Extrapolation of those leak rates into the future produces bounding estimates for total inventory losses for the duration of each leak. Therefore, it is important to consider the causes for tank leaks in order to better determine their potential start dates.

Leaks often appear on the tails of tank heat load curves. It is not clear at this time if this is because there was some physical mechanism that took place with tanks during these periods or if that means that the high reflux rates for these tanks prevented any effective determination of unaccounted volume losses. The evaporation rate for a tank's waste is an increasingly sensitive function of the tank temperature and ventilation rate during these times. The HLM assumes that the tank heat loss to ground and air are constant during these times at 10 kW each for a total loss of 20 kW.

Bounding estimates for leaks from waste tanks depend critically on the timing of a leak as well as on its size. If a leak begins during times of high thermal stress, which is a reasonable assumption, it will be masked by the correspondingly large reflux rates and most likely will not be evident until the reflux rate of a tank is reduced considerably . Only then will the leak rate become a significant fraction of the normal makeup volume. Moreover, tanks connected to a common ventilation system will be subject to the reflux rate of the entire system, not just that for individual tanks. This interlinking of the tanks would have seriously complicated any determination of volume unaccounted for an individual tank.

Reasons for tank failure:
Stress-corrosion cracking failure has been most often blamed for the majority of tank leaks. It begins at welds that have residual stress and propagates as cracks at right angles to the welds. Thermal stress presumably contributes to increasing these corrosion rates and many single-shell tanks were subjected to temperatures on the order of 140-160°C (280-320°F) for sometimes ten or more years. Newer double-shell tanks were stress relieved prior to use and none have shown any sign of leaking. On the other hand, the DST's have not been subjected to the very high thermal stresses and severe steam bumps that were common for SST's in S, SX, A, and AX Farms.

Undoubtedly stress-corrosion cracking has played a major role in many tank leaks, but high thermal stresses leading to sudden structural failure cannot be neglected as a potential explanation for tank leaks for SST's with large thermal loads. This mode of failure is certain for at least three tanks at Hanford: SX-108, SX-113, and A-105. Severe bottom bulges were observed for each of these tanks and directly associated with their leaking waste into the soil column. For both SX-108 and SX-113, the bottoms relaxed back to normal some years after they bulged. It is very likely therefore that other high heat tanks also suffered bottom bulging that also relaxed back and therefore were never observed. Some eleven other leaker tanks that were thermally stressed are: A-103, A-104, S-104, SX-104, SX-107, SX-109, SX-110, SX-111, SX-112, SX-114, and SX-115. In fact, bent temperature probes, which are highly suggestive of past deformation of a tank bottom, are present in two of these tanks: A-104 and SX-109.

It is clear then that sudden structural failure at least played a role in tank failures at Hanford. The question becomes how can we use that information to help define the tank leak amount and timing.

Another factor to consider is the total volume of soil associated with the leak. Drywells are positioned 10' from the tank edge and for tank SX-108, 08-11 and 08-02 both show activity centered on 55' depth. Assuming that the entire soil quadrant is saturated to 10' from the outer edge of the tank, this would require 4.4 kgal waste per foot of saturation depth with a 20% porosity of the soil. Thus, a 203 kgal leak should have a saturation depth of 46' .

Leak Model:
Each leak is described by three parameters, a leak elevation (in equivalent kgal tank volume), a leak rate (in kgal leaked per month per kgal head above the leak elevation), and a leak period. Thus, a leak is assumed to derive from a particular elevation within a tank and therefore a tank only leaks when the waste level is greater than the leak elevation. The leak rate then increases linearly

as the hydraulic head above the leak elevation increases. Finally, each leak has a starting time and a duration. Note that since tank waste elevation and hydraulic head drive the leak rate, the resultant leak rate may vary substantially over its duration depending on the changes in the waste level within the tank.

A. $\text{leakVol}(\text{mo}_i) = \text{leakSize} * (\text{calcVol} - \text{leakElev})$

B. $\text{leakAcc} = \Sigma_i \, \text{leakVol}(\text{mo}_i)$

In addition, there are numerous reports of leaks that “self-sealed” after they had begun and then perhaps later restarted. As a result, another potential variable is the leak “duty-factor” or that fraction of time that a leak actually occurred over the entire leak duration. However, there is simply not enough information for all these tanks to justify yet another leak parameter and so leaks for SX-108, SX-109, and SX-111 will be assumed to be continuous for their duration with a rate dependent only on tank elevation and hydraulic head.

For tank SX-112, there is a period of unaccounted loss in 1958-9 that suggests a loss of around 1.6 kgal/mo for 12 months. This is followed by a second cooling period 1964-66 that did not show unaccounted losses and then a final cooling period in 1969 where unaccounted losses occured once again. As a result, we have included two separate leaks for this particular tank with the assumption that the leak effectively sealed in between these events.

The difference in slope between the two lines (calc. vol. and vol w/o leak) for tank SX-108 (Fig. A-1) from 1960 to 1962 represents the leak rate for this tank. In other words, the tank heat load at this time was not sufficient to explain the reported waste volume losses. Likewise, these differences in slope define leaks for SX-109 (1967-9) and SX-111 (1973-4). For tank SX-112, there is much more limited data for its second leak and that leak is defined by only two months in 1969.

Soluble radionuclides are assumed to all follow Cs-137 and this is the only isotope calcuted by the HLM. All other radionuclides are calculated by scaling results from the HDW model estimates (see below).

Reflux model:
During the reflux process, each tank’s dome vapor was routed by an underground header system to a single or centralized condenser. Water was extracted from the tank dome vapor down to the dew point of the condenser temperature, which we presume was on the order of 65°F (18°C). However, this condenser temperature does not affect the HLM. The chilled air was then either vented to the atmosphere or returned to each tank. The condensate from this process was then returned to each tank as needed to maintain some target level for that tank along with some makeup volume. However, since it was often the case that many tanks were connected to a common ventilation system, the exact amount that needed to be returned to each tank could only be determined by the losses that were observed for that tank.

This meant that if some other loss pathway existed for the tank waste, for example a leak, this volume would also need to be made up. By measuring the makeup volume needed over and above the condensate extracted, one could then surmise if there were losses over and above those expected from makeup volume and take appropriate action to isolate which tank was leaking. Unfortunately, the normal steam vapor losses for the system must have scaled as something like the total reflux volume and these losses needed to be made up as well during the process. As a result, the normal makeup volume for the total system undoubtedly masked many individual tank leaks to the soil column.

Since the level record of each tank is mostly expressed quarterly or semiannually, there are substantial gaps in the volume record and those gaps would need to be filled to actually simulate each tank adequately. In particular, we do not believe that the tank waste volumes decreased to the extent represented by the HLM over a period of a quarter. The HLM does show the intermediate volumes decreasing substantially during many quarters and we presume that the actual decrease was limited to some more reasonable amount. For example, during 1956-60 we have monthly reports for these tank levels as well as monthly reports of concentration and water additions.

The amount of waste evaporated was calculated as

C. $\quad evapRate\ (in/mo_i) = tankHeat(kW)\ /\ waterHeatOfVap(kW/(in/mo))$

where

D. $\quad waterHeatOfVap = 8.96\ kW/(in/mo)$
(from heatVap = 2259 J/g x 3.785e6 g/kgal x 2.75 kgal/in / 2.625e6 s/mo)

is simply that of pure water at 100 C. The spreadsheet calculated total volume in kgal for each tank was

E. $$calcVol(mo_i) = calcVol(mo_{i-1}) + tankTrans(mo_i) - 2.75*evapRate(in/mo_i) + accWater(mo_i) + unaccWater(mo_i)$$

where there are 2.75 kgal/in and unaccounted water addition was based on

F. $$unaccWater(mo_i) = measVol(mo_i) - - (measVol(mo_{i-1}) + tankTrans(mo_{i-1}) + accWater(mo_{i-1}) + unaccWater(mo_{i-1}) - evapVol(mo_{i-1}) - leakVol(mo_{i-1})).$$

Likewise, the tank volume in the absence of a leak was simply

G. $$noLeakVol(mo_i) = calcVol(mo_i) + leakVol(mo_i).$$

Note that calcVol and measVol coincide by adjustment of unaccWater in an iterative spreadsheet calculation. Thus, any changes in leak or other parameters cause the spreadsheet to recalculate to a new set of unaccWater additions for that tank. This calculation takes several minutes and is iterative since changes in unaccWater for a given month affect all subsequent months.

IV. Results:

Tank leak estimates are primarily derived by examination of unaccounted volume losses, but we use the lateral and drywell contamination reports to corroborate leak start times as well. We attribute all unaccounted volume gains to water additions that replace each tank's evaporative loss.

Tanks in SX Farm were cooled primarily by reflux during this time. The reflux involved extraction of steam from tank dome space followed by contact with a condenser to condense and remove water. The condensate from this process was returned to the tank and the air was either vented to the atmosphere or returned to the tank. Since tanks with very high reflux rates lost tank inventory at very high rates (up to 200 kgal/mo), there were correspondingly large uncertainties in unaccounted volume losses associated with this reflux cooling of each tank. Most of the inventory loss during reflux was immediately returned to the tank as water, but some undoubtedly also escaped as steam during the operation. This condensate loss had to be made up by periodic water additions and the amount of this makeup could effectively mask relatively small losses due to leaks.

Many tanks were connected to a common ventilation system in SX Farm which would have further complicated the already difficult determination of material balance. A tank that developed a leak contributed an unaccounted volume loss to an entire group of tanks that were on a common ventilation system. Therefore, a leak could be masked not only by its own high evaporation rate but also by other tanks within a group with high boil-off rate tanks.

We know that 242-T and 242-S in-farm evaporators used a 5% materials accountability criterion for their operation [see evaporator references]. Because the in-tank evaporation for SX Farm was much more complex than the in-farm evaporators, this suggests that SX Farm condenser operation probably used a materials balance on the order of 5-10%.

Table 2. Comparisons of HLM with other leaks for SX-108, SX-109, SX-111, and SX-112.

Tank	Hanlon Date	Hanlon kgal	Anderson Date	HLM Date	HLM kgal
SX-108	1962	35	Mar. 1959	Jun. 1959	203
SX-109	1965	10	Dec. 1967	1960	111
SX-111	1974	2	Jun. 1974	Jun. 1972	55
SX-112	1969	30	Mar. 1969	Oct. 1958, Mar. 1969	44

Hanlon is Hanlon, B.M. "Waste Tank Summary Report of Month Ending May 31, 1996," WHC-EP-0182-99, August 1996.
Anderson is Anderson, J.D. "A History of the 200 Area Tank Farms," WHC-MR-0132, June 1980.

Table 3. HLM leaks for SX-108, SX-109, SX-111, and SX-112.

tank	leak start	leak end	leak rate (kgal/mo/kgal head)	leak elevation (kgal)	leak volume (kgal)	leak Cs-137 MCi*
SX-108	1959.5	1967.5	0.0055	200	203	0.43
lower					102	
SX-109	1960.0	1969.7	0.0025	200	111	0.32
lower					56	
SX-111	1972.5	1975.0	0.0035	200	55	0.009
lower					14	
SX-112	1969.0	1970.0	0.0025, 0.05	200	44	0.25
lower					22	
totals **lower**					413 kgal 194	1.01 MCi
Hanlon					73 kgal	

*decayed to 1/1/94

Notes on specific tanks:
SX-108
Tank SX-108 showed its first HLM unaccounted volume loss in 1959 (see App. A, column "unacc.vol.", third quarter 1959). The HLM leak assignment begins at this time as shown in Fig. A1 and column "kgal leak". A bottom bulge may have appeared as early as 1959 but a tank bottom bulge was reported in September 1967 and subsequently relaxed back to a flat position [Brevick, et al.]. There are bent temperature probes in the northwest quadrant of this tank yet today, which is about where the bulge was reported previously. The drywell with the greatest activity is 08-11, which is adjacent to the northwest tank quadrant, once again the same quadrant that bulged. In addition, all three lateral wells underneath this tank show large amounts of activity.

Although there were reports of lateral well activity in SX-108 as early as December 1962, the tank was emptied and refilled with another round of fresh Redox waste from mid 1962 through 1964.

This tank was placed in reflux during this time with a peak evaporation rate by the HLM of 132 kgal/month (48 in/mo of waste inventory) in late 1963. By mid 1966, the reflux rate of the tank had slowed significantly and the tank once again showed a steady loss in volume over what was expected based on its HLM heat load.

Concern with increased lateral well activity in November 1965 dictated that the tank be placed on an isolated cooler and over the next several months, the tank's level was reported to have stabilized and the leak therefore assumed to have "self-sealed." The level data within the HLM, though, shows no such level stability at that time. In fact, the HLM SX-108 inventory decreased 13 kgal from the December 1965 of 653 kgal to the June 1966 level of 640 kgal. The reflux rate predicted by the HLM in November 1965 was 20 kgal/mo while the HLM leak rate at that time was 1.6 kgal/mo. It is not clear that such a leak rate would have yet been evident for this tank given this rate of boil-off and so a continuing leak is possible during this time despite the reports of the tank's leak having "self-sealed."

Maximal and minimal leak volumes are derived by assuming a continuous leak with the leak parameters shown in Table 3 (maximum leak rate was ~3 kgal/mo and varied according to waste level). Total volume loss up through 1962 was 203 kgal if the leak is assumed to have continued through 1967. These leak amounts depend critically on assumptions of heat losses to ground and air as well as on the continuous nature of the leak. Therefore, each leak estimate may be ~50% lower due to inaccuracies in the HLM for low heat load tanks. No account is taken in this analysis for potential "self-sealing" or non-continuous leaks. Unaccounted volume losses after 1967 may have been another leak or simply an extension of the old one, but since the waste within the tank was solidifying at that time, these level changes become increasingly difficult to interpret. Considering the fact that some inventory in the tank seems to spontaneously appear in 1969, we attribute this later unaccounted volume loss to a measurement anomaly and accordingly ignore it.

A leak volume of 203 kgal represents 27,100 cu.ft. of waste which would wet approximately 135,000 cu.ft. of soil at 20% porosity. This volume of soil is a sphere approximately 64' in diameter and therefore would imply a lateral penetration into the soil column underneath the tank on the order of 32'.

Figure A2 compares the rates for unaccounted volume gains and losses with those for leak and evaporation for the period 1959 to 1962. We derived the leak rate for SX-108 by adjusting it to accommodate as much of the unaccounted volume losses as reasonable. This produces the leak rate shown in Fig. A2 as well as a set of unaccounted volumes that now represent no change in volume or volume gains (solid diamonds). Fourth quarter 1960 still shows an unaccounted volume loss, but we felt that this quarter's large unaccounted volume loss was beyond that that the HLM could reasonably accommodate. This volume loss may represent a variable leak rate or it may represent the uncertainty of data within the HLM.

SX-109

Tank SX-109 shows its first unaccounted volume loss in 1960 whereas Hanlon reports 1965, Anderson reports December 1967, and other sources report January 1965. Significant long term unaccounted volume losses continued for many years for this tank. The HLM shows this time period to have had insufficient heat load to explain all long-term volume loss by evaporation. These unaccounted volume losses amount to 111 kgal over these many years. This amount may once again be as much as 50% too large because of HLM inaccuracies for low heat loads. Thus, a lower bounds would be 56 kgal leak for this tank. This is a tank that shows clear evidence of a very slow leak over a long period of time, which is consistent with the fact that the drywells associated with this tank were showing readings consistent with continued migration of radionuclides as late as 1976. There are bent temperature probes in the north edge and southeast quadrant of this tank but no bottom bulge was ever reported for SX-109. The drywell 09-04 that is adjacent to the southeast quadrant does show activity associated with a leak.

Figure A4 compares various volume rates for SX-109. The leak rate for this tank was adjusted to accommodate all unaccounted volume losses. Note that the leak rate reduces significantly in late 1960 as a result of the fact that leak rates scale with tank volume and tank volume changed at this time (see Fig. A3).

SX-111
Tank SX-111 shows an HLM unaccounted volume loss in third quarter 1972 of about 2.4 kgal/mo. Extending this leak until the tank was pumped in second quarter 1974 amounted to a total of 55 kgal for this leak duration. Since this unaccounted volume loss appeared at a time of very low heat load, it is subject to increased uncertainty because of the HLM deficiency with low heat loads in tanks. During the period of the leak, the evaporation rate decreased only slightly from 5.5 to 5.0 kgal/mo while that of the reflux coupled system dropped from 15 to 12 kgal/mo. Thus, this leak estimate may be as much as a factor of four too large, giving a 15 kgal lower limit for this leak.

At any event, lateral and ground contamination in 1974 resulted in this tank being assigned as a leaker. Tank SX-111 has three other periods in the tails of cooling curves where a leak should show up; one in 1958-9, the second in 1963-4, and a third in 1969-71. Neither of these periods show indication of unaccounted volume losses by the HLM, which suggests that the HLM is reasonably well calibrated. Figure A6 shows the comparison of volume rates for SX-111 for the period 1972 through 1974. Since there is only one quarter showing an unaccounted volume loss there is a great deal of uncertainty is associated with this leak assignment.

SX-112
Tank SX-112 shows its first HLM unaccounted volume loss in October 1958 with several additional months unaccounted losses in 1959 as well. At this time, boil-off was down to 1-2 kgal/mo while the unaccounted losses ranged up to 4 kgal/mo, suggesting a leak of around 1.6 kga;/mo over 12 months for a total leak volume of 19 kgal. These adjustments in unaccounted volume are shown in Figs. A8 and A9.

However, this tank was only noted as a leaker in 1969 as shown in Figs. A9, which is consistent with the assignment of this tank as a leaker by other sources. Figure A9 shows the volume rates for SX-112 where we have only a single quarter of unaccounted volume loss, first quarter 1969. This tank was pumped to heel in the second quarter and therefore we have a very limited duration for this leak with a loss of 25 kgal over 2 months or about 13 kgal/mo. At that time, SX-112 was evaporating at the rate of 16 kgal/mo while the system rate reflux was 50 kgal/mo. Since this leak was detected very soon after the unaccounted volume loss, we assume that the drywell contamination was evident at this time as well. Currently, we estimate that the total amount leaks was 44 kgal from both leak periods but that this may be as much as 50% too high or low as a result of the possibility of variation of either leak period.

SX-105
Tank SX-105 has not been assigned as a leaking tank and it is the only SX Farm waste tank outside of the leaking set, SX-107 through SX-115, that has lateral as well as vertical drywells. Therefore, it provides an ideal non-leaking tank with which to test the limits of the HLM. Application of the HLM to this tank is shown in Figs. A10 and A11. The 1957-63 cooling period shows only three calendar quarters of very slight unaccounted volume losses of 0.5 kgal/mo or less as shown in Fig. A11. This size of unaccounted loss represents the limit of leak that the HLM can discern. This leak rate corresponds to a leak size of 7.6e-4 kgal/mo per kgal head, or about a factor of three less that for SX-109 and a factor of six less than the leak sizes for SX-108, SX-111, and SX-112. We assume that the HLM simply is not sensitive enough to discern leaks of this size.

V. Description of HLM Spreadsheets
Appendix A shows the plots of the fill, thermal, and heat load histories of these tanks as well as the spreadsheets that were used to generate the plots. Also shown in each plot is the calculated volume of each tank in the absence of any leak. The criteria used to define each leak basically

depended on the comparing of unaccounted volume loss rates and adjusting the size and duration of each leak in order to reduce those unaccounted volume losses.

Figures A1, A3, A5, A7, and A10 shows reported tank volumes, calculated tank volumes with leak, calculated tank volume without leak, calculated heat load of the tank, and reported tank temperature. All of this information is shown on a month-by-month basis in the HLM although much of the historical information that we now have is tabulated quarterly and sometimes only semi-annually.

Figures A2, A4, A6, A8, A9, and A11 all show the volume rates for evaporation, leak, and unaccounted gain or loss. These plots illustrate the means of calibrating the leaks using unaccounted volume loss information.

VI. Leak inventories

Given the leak volumes, starting times, and ending times, we calculate the composition leaked in terms of R1, R2, RSltCk, and other HDW types. Given the composition of each of these wastes, we then derive leak inventories for each chemical and radionuclide. As tank wastes were concentrated and blended, the compositions changed somewhat from the original wastes that were placed in each tank. These changes have been calculated by the SMM (Supernatant Mixing Model), which assumes an initial sludge/supernatant partitioning and propagates partitions of each waste type expressed in kgal's of original waste.

We also calculate the amount of Cs-137 placed into each waste tank directly from fuel element processing records. This provides a parallel estimate of the Cs-137 that leaked from each of these tanks independent of the blending approximations inherent within the HDW model. These estimates are shown in Table 5 in total MCi (1 MCi = 1e6 Ci = 3.7e4 Tbq). The difference in the HLM and HDW model's Cs-137 totals is due to the approximations of the HDW model. The HDW model defines wastes that are blended composites of entire campaigns whereas the HLM has placed each batch of fuel into tanks on a month-by-month basis. Therefore, the soluble radionuclides should all be corrected with the factors shown in Table 5.

For example, Tc-99 for tank SX-108 is 1.07e-4 Ci/L, corresponding to a leak inventory of 203 kgal x 3785 L/kgal x 1.07e-4 = 82 Ci Tc-99 by the HDW estimate. For the HLM estimate, multiply by 1.6 to get 131 Ci Tc-99. This assumes that Tc-99 behaves like Cs-137 in the tank liquors.

Table 4. Chemical and Physical Characteristics of HLM Leaks.

mol/L	SX-108	SX-109	SX-111	SX-112
Na	7.55E+00	5.39E+00	3.41E+00	6.32E+00
Al(OH)4 -	1.51E+00	1.14E+00	3.77E-01	1.60E+00
Fe	4.30E-03	2.77E-03	4.15E-03	2.77E-03
Cr	2.58E-01	1.66E-01	8.96E-01	1.66E-01
Bi	1.29E-06	1.88E-06	9.00E-05	2.51E-06
La	5.01E-12	7.27E-12	1.69E-09	9.70E-12
Hg	2.03E-07	2.95E-07	9.82E-07	3.94E-07
Zr	6.28E-07	9.11E-07	5.18E-05	1.22E-06
Pb	3.22E-05	4.68E-05	1.32E-04	6.24E-05
Ni	3.87E-03	2.49E-03	1.42E-03	2.49E-03
Sr	1.67E-12	2.42E-12	5.64E-10	3.23E-12
Mn	9.42E-06	1.37E-05	2.85E-03	1.82E-05
Ca	1.93E-02	1.25E-02	7.11E-03	1.24E-02
K	3.07E-02	2.25E-02	1.69E-02	2.98E-02
density g/cc	1.39	1.28	1.15	1.35
wt.% H2O	59.4	68.9	78.8	66.7
TOC wt.%C	1.22E-03	1.92E-03	3.17E-01	2.43E-03
species				
OH-	7.04E-02	4.62E-02	3.49E-01	3.78E-02
NO3-	3.57E+00	2.41E+00	1.15E+00	2.20E+00
NO2-	2.34E+00	1.73E+00	9.27E-01	2.34E+00
CO3--	2.03E-02	1.38E-02	1.68E-01	1.43E-02
PO4---	8.35E-05	1.21E-04	1.01E-02	1.62E-04
SO4--	5.18E-02	3.78E-02	9.72E-02	4.75E-02
SiO3--	4.51E-02	3.24E-02	2.68E-02	4.68E-02
F-	7.40E-05	1.07E-04	4.55E-03	1.43E-04
Cl-	1.41E-01	1.03E-01	6.02E-02	1.37E-01
C6H5O7---	6.90E-05	1.00E-04	2.08E-02	1.34E-04
EDTA----	2.69E-06	3.90E-06	3.72E-04	5.20E-06
HEDTA---	2.23E-06	3.24E-06	7.53E-04	4.32E-06
glycolate-	9.74E-05	1.41E-04	4.79E-03	1.89E-04
acetate-	1.01E-05	1.47E-05	2.48E-06	1.96E-05
oxalate--	4.17E-12	6.06E-12	1.41E-09	8.09E-12
DBP	6.12E-05	8.88E-05	1.32E-02	1.18E-04
butanol	6.12E-05	8.88E-05	1.32E-02	1.18E-04
NH3	4.80E-02	3.65E-02	2.68E-02	5.92E-02

Table 5. Radionuclide Concentrations for HLM Leaks.*

Ci/L	SX-108	SX-109	SX-111	SX-112
Sr-90 (Ci/L)	7.31E-02	4.71E-02	5.28E-02	4.70E-02
Tc-99 (Ci/L)	1.07E-04	8.17E-05	1.47E-04	1.28E-04
I-129 (Ci/L)	2.00E-07	1.53E-07	2.79E-07	2.38E-07
Cs-137 (Ci/L)	3.49E-01	2.71E-01	4.54E-02	4.44E-01
U-232 (Ci/L)	2.35E-09	3.39E-09	6.24E-08	4.53E-09
U-233 (Ci/L)	8.76E-09	1.27E-08	2.38E-07	1.70E-08
U-234 (Ci/L)	5.15E-07	3.50E-07	2.63E-07	3.71E-07
U-235 (Ci/L)	2.13E-08	1.43E-08	1.09E-08	1.43E-08
U-236 (Ci/L)	1.56E-08	1.21E-08	6.96E-09	2.15E-08
U-238 (Ci/L)	4.76E-07	3.15E-07	2.55E-07	2.80E-07
U-Total (mol/L)	5.98E-03	3.95E-03	3.14E-03	3.50E-03
Np-237 (Ci/L)	5.89E-07	4.47E-07	9.08E-07	6.73E-07
Pu-238 (Ci/L)	4.40E-07	3.19E-07	4.95E-07	5.50E-07
Pu-239 (Ci/L)	1.90E-05	1.22E-05	2.31E-05	1.21E-05
Pu-240 (Ci/L)	2.79E-06	1.85E-06	3.49E-06	2.15E-06
Pu-241 (Ci/L)	2.28E-05	1.64E-05	3.43E-05	2.68E-05
Pu-242 (Ci/L)	1.16E-10	8.58E-11	1.42E-10	1.55E-10
Pu-Total (g/L)	3.18E-04	2.05E-04	2.20E-04	2.05E-04
Am-241 (Ci/L)	3.69E-05	2.98E-05	6.21E-05	5.42E-05
Am-243 (Ci/L)	1.31E-09	1.15E-09	1.93E-09	2.47E-09
1 MCi = 1e6 Ci				
HDW Cs-137 MCi	2.68E-01	1.14E-01	1.06E-02	9.62E-02
HLM Cs-137 MCi	4.25E-01	3.21E-01	9.0E-03	2.5E-01
soluble. nuclide correction	1.6	2.8	0.93	2.6

*Decayed to 1/1/94

Uses and Limitations:
Clearly, there are limitations to the HLM for leak determination and there are also limitations in every approach that has been used in the past for leak determination. The HLM is only appropriate for aging waste tanks where water evaporation is cooling the tank waste. This limits its use to SX, A, AX, and possibly S and BY farms as well. Second, the HLM is only able to discern leak rates on the order of 0.5 kgal/mo or greater and it really is meant to record trends of volume lost over many months of operation.

The HLM has assumed a constant conductive heat loss for each tank that is scaled with tank temperature relative to seasonal average ground and air temperatures. Therefore, seasonal variations in temperature are not taken into account and these are appreciable for heat loss to air. Seasonal variation of ground temperatures is much less and therefore not expected to be a substantial source of HLM variation. Likewise, diurnal variations in temperature are not expected to have an impact on tank heat loss variation. The waste tanks are very large and therefore simply do not respond to temperature variations on the time scale of one day.

These four tanks were chosen for the HLM analysis because they had already demonstrated a substantial amount of contamination to the soil column by their associated drywell activities. The real question for these tanks was not if they leaked, but rather how much they leaked. The HLM confirms that these tanks have unaccounted volume losses and uses those unaccounted losses to extend the tank leaks over periods of operation where leak determination was very difficult by any other means.

The partitioning of radionuclides among tanks by subsequent transfers of waste supernatant is modeled only very crudely by the HLM. We have not evaluated the effect of the partitioning assumption on the leak estimates.

Acronyms, Abbreviations, and Definitions

kgal	1,000 gallons
MCi	1e6 (one million) Curies = 37,000 TBq, 1 Ci = 3.7e10 Bq
HLM	Historical Leak Model
Redox	Reduction and Oxidation solvent extraction for Hanford.
Sr	Strontium
Cs	Cesium
SX Farm	Tank farm in 200 West Area at Hanford Site in Washington State
A, AX Farms	Tank farms in 200 East Areas at Hanford Site in Washington State
CRS	Tank Advisory Panel Chemical Reactions SubPanel
WSTRS	Waste Status and Transaction Record Summary
ORIGEN2	Computer code developed at Oak Ridge National Laboratory for predicting fission products from reactor fuel burnup.
HDW	Hanford Defined Wastes model uses process histories to predict the contents of Hanford waste tanks.
Excel	Registered trademark of spreadsheet software from Microsoft.
S Plant	Another name for the Redox plant.
measVol(mo_i)	Reported tank level by direct measurement.
calcVol	Calculated Tank volume in kgal calculated based on previous month, quarter, or biquarter with recorded transactions, evaporation based on heat load, and leak.
leakVol(mo_i)	Volume rate in kgal/mo for leak.
leakSize	Leak size parameter in kgal/mo per kgal head above leak elevation.
leakElev	Leak elevation is actually set to 200 kgal elevation in tank. This scales all leaks to the same tank elevation.
leakAcc	The total accumulated leak volume in kgal.
evapRate (in/mo_i)	Evaporation rate in in/mo.
evapVol(mo_{i-1})	Volume in kgal evaporated for month i.
tankHeat(kW)	Tank heat load in kW.
waterHeatOfVap	8.96 kW/(in/mo) (from heatVap = 2259 J/g x 3.785e6 g/kgal x 2.75 kgal/in / 2.625e6 s/mo)
tankTrans(mo_i)	Volume of transaction for month i.
unaccWater(mo_i)	Volume of unaccounted water gain or loss for month i.
accWater(mo_{i-1})	Accounted water as condensate reported for month i.
noLeakVol(mo_i)	Tank volume in kgal with leak volume added back in.

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Appendix A.
Historical Leak Model Figures and Spreadsheet

The HLM spreadsheets contain the heat loads, evaporation rates, unaccounted volume losses, and tank levels for each of four tanks: SX-108, SX-109, SX-111, and SX-112. In addition, tank SX-105 volume history is shown as a comparable non-leaking tank. These spreadsheets are interlinked with each other electronically as well as with radionuclide source spreadsheets. The unaccounted volume losses and their dates are generated from this data and serve as the key information that defines leaks within the HLM.

The charts summarize either volume or volume rate data for each tank and Table A1 describes the column headings and corresponding spreadsheet equations.

Table A1. Description of columns and formulas.

col #	label	description	spreadsheet formulas (B1 is prior row, column 1; C23 is same row, column 23)
1	**tank label**	fractional year	=$B1+1/12
2	mo.	calendar month	3
3	meas. vol.	reported volume of tank in kgal	634
4	calc. vol.	calculated tank volume based on last month less evaporation plus transfers less leak.	=$B4+$C5-$C23*2.75+$C24+$C25-$C50
5	kgal added	volume of accounted waste transfers (positive for waste additions, negative for waste removals)	
6	dwxt	HDW definition of waste type added	
7	fr. rads.	fraction of radionuclides processed that month that are added to tank.	
8	MCi Sr-90	total megacuries of Sr-90 in tank decayed to that time.	=$C43/'red-u3'!$U1/1000000
9	MCi Cs-137	total megacuries of Cs-137 in tank decayed to that time.	=$C33/'red-u3'!$N1/1000000
10	waste age	[future use]	
11	kgal vol.	[future use]	
12	kgal sl	[future use]	
13	kW sl	[future use]	
14	kW su	[future use]	
15	kW total	calculated heat load based on radionuclides	=($C29+$C30+$C31+$C32+$C33+$C39+$C40+$C41+$C42+$C43)/1000
16	tank temp. deg.C	tank waste temperature. If not available, assumed 120 C for boiling waste, 80 C otherwise.	129
17	ground temp. deg. C	assumed seasonal average	18
18	air temp. deg.C	assumed seasonal average	19
19	vent. rate cfm	assumed constant	100
20	kW to ground	calculated heat conduction to ground	=($C16-$C17)*rad.heating!$Q$2
21	kW to air	calculated heat conduction to air	=($C16-$C18)*$C19*rad.heating!$Q$3
22	kW to evap.	kW heat lost to evaporation [not used]	
23	evap. rate in/mo.	calculated evaporation rate (kW total less kW to ground and kW to air) divided by volumetric heat of vaporization for water.	=IF(($C15-$C20-$C21)/rad.heating!$O4<0,0,($C15-$C20-$C21)/rad.heating!$O4)
24	acc. water	accounted additions of water in kgal	
25	unacc. vol.	unaccounted additions of volume in kgal (negative if unaccounted loss)	=$C3-(B3+$A5+$B5+$C5+$C24+$B24+$A24-($A23+$B23+$C23)*2.75-($A50+$B50+$C50))
26	acc. cond.	kgal accounted additions of condensate to tank	
27	kgal evap.	calculated kgal evaporated based on evap.rate	=-$C23*2.75
28	fr. lost	fraction of heat load lost that month with volume lost from tank, either by leak or transfer.	=IF($C5<0,(-$C5+$C50)/$B4,$C50/$B4)
29	C1 W	accumulated C1 isotope inventory in watts	=($B29*EXP(-LN(2)/'red-

			u3'!$N$10/12)+$C34)*(1-$C$28)
30	C2 W	accumulated C2 isotope inventory in watts	=($B30*EXP(-LN(2)/'red-u3'!$N$8/12)+$C35)*(1-$C$28)
31	C3 W	accumulated C3 isotope inventory in watts	=($B31*EXP(-LN(2)/'red-u3'!$N$6/12)+$C36)*(1-$C$28)
32	C4 W	accumulated C4 isotope inventory in watts	=($B32*EXP(-LN(2)/'red-u3'!$N$4/12)+$C37)*(1-$C$28)
33	Cs-137 W	accumulated Cs-137 isotope inventory in watts	=($B33*EXP(-LN(2)/'red-u3'!$N$2/12)+$C38)*(1-$C$28)
34	C1 W in	addition of C1 isotope watts with waste input	
35	C2 W in	addition of C2 isotope watts with waste input	
36	C2 W in	addition of C3 isotope watts with waste input	
37	C4 W in	addition of C4 isotope watts with waste input	
38	Cs W in	addition of Cs-137 watts associated with waste input for this month.	
39	S1 W	accumulated S1 isotope inventory in watts	=$B39*EXP(-LN(2)/'red-u3'!$U$10/12)+$C44
40	S2 W	accumulated S2 isotope inventory in watts	=$B40*EXP(-LN(2)/'red-u3'!$U$8/12)+$C45
41	S3 W	accumulated S3 isotope inventory in watts	=$B41*EXP(-LN(2)/'red-u3'!$U$6/12)+$C46
42	S4 W	accumulated S4 isotope inventory in watts	=$B42*EXP(-LN(2)/'red-u3'!$U$4/12)+$C47
43	Sr-90 W	accumulated Sr-90 isotope inventory in watts	=$B43*EXP(-LN(2)/'red-u3'!$U$2/12)+$C48
44	S1 W in	addition of S1 isotope watts with waste input	
45	S2 W in	addition of S2 isotope watts with waste input	
46	S3 W in	addition of S3 isotope watts with waste input	
47	S4 W in	addition of S4 isotope watts with waste input	
48	Sr W in	addition of Sr-90 watts associated with waste input for this month.	
49	heat load	total heat load in millions of Btu/hr	=$C15/0.293/1000
50	kgal leak	kgal leak for month	=IF(AND($C$4>$BA$2,$C$1>$BA$3,$C$1<$BA$4),($C$4-$BA$2)*$BA$1,0)
51	accum leak	kgal accumulated leak	=$B51+$C50
52	leak rate leak elevation leak start leak end	leak rate as kgal per month per kgal of head above leak elevation; elevation of leak in tank; fractional year for start of leak; fractional year for end of leak.	
53			
54	w/o leak	calculated tank waste plus leak volume	=$C4+$C51
55	temp	temperature of tank in C (same as column 16)	129
56	unacc.loss	unaccounted volume loss remaining after leak correction.	=IF($C25<0,$C3-$C25,10000)

Appendix B.
Overview of Redox Process

The Redox process was based on the extraction or salting out of plutonium and uranium from an aqueous aluminum nitrate solution into an organic phase, methyl isobutyl ketone also known as hexone (see Fig. B1). Anderson-91 describes the various stages in the development of the Redox process, which began in January 1952 at S or Redox Plant.

According to Anderson, waste was originally generated at 4,378 gal/ton in 1952, and that rate was reduced to 594 gal/ton in 1966. We have found by analyzing the fill records, on the other hand, that the waste rate peaked at around 4,600 gal/ton in 1952 and after around 1958, leveled off to around 1,100 gal/ton.

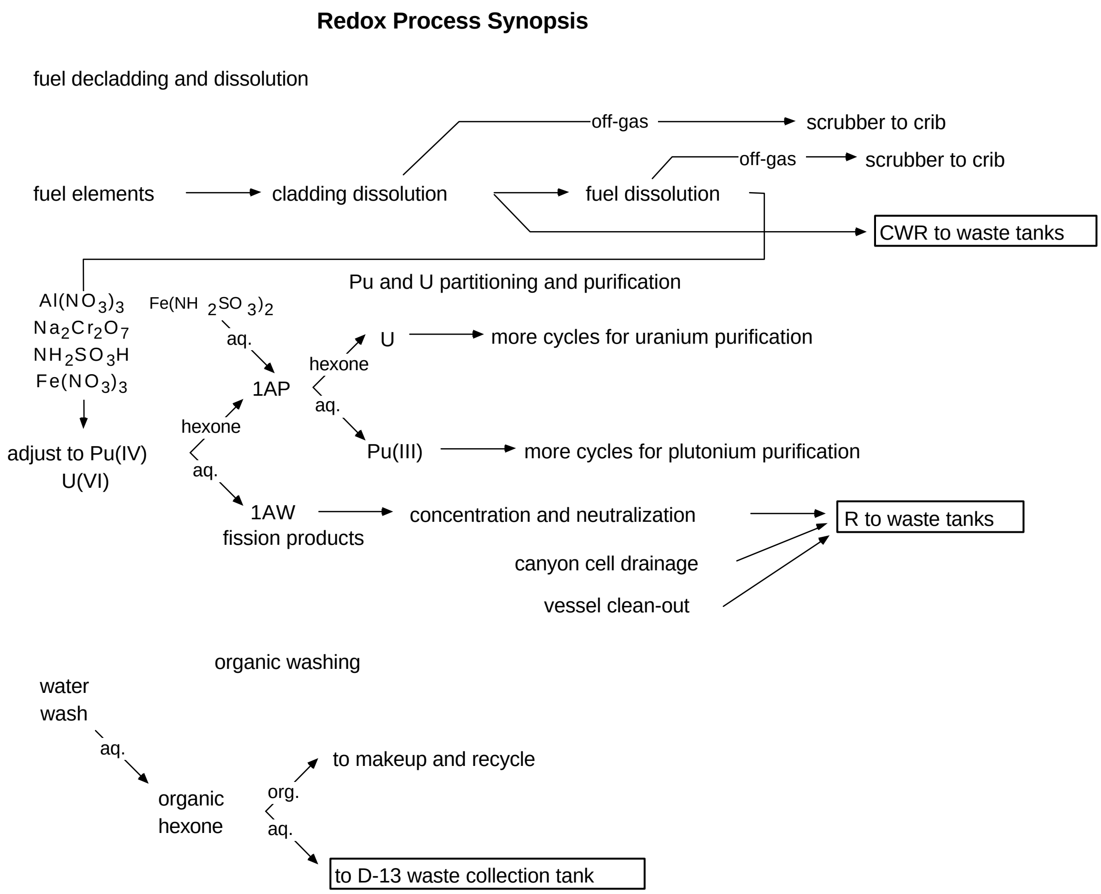

Figure B1. *Redox process synopsis.*

Thus, there were essentially two eras for the Redox waste, the first era from 1952-58 averaged 2,106 gal/ton, followed by a reduction to 1,119 gal/ton from 1959-66 (see Fig. B3). We do find a waste rate as low as 500 gal/ton in the last quarter of 1966, but averaged for all 1966, the last full year of operation, Redox generated waste at the rate of 1,085 gal/ton.

We have also found that the cladding waste generation rate (CWR) was fairly constant at 266±30 gal/ton over the entire history of Redox, as opposed to a remark by Anderson-91, that cladding waste volumes were cut in half in 1956-57. We have found no such decrease in CWR waste rates averaged for any year of operation over the entire Redox campaign. There are some 980 kgal of CWR that is reported by WSTRS after all fuel was no longer processed in Redox in mid 1966 (see

Fig. B3). We assume that the fuel slugs from this decladding operation were actually processed in Purex Plant.

The fuel slugs were coated with a bronze layer (Cu and Sn) prior to being welded in their jackets. Neither of these elements are currently within the HDW chemicals added and so are not included in the cladding waste estimates. Moreover, there are other reports of lead dips being used instead of bronze for some fuel slugs and we have included lead in the cladding waste chemicals added definition.

Anderson also mentions that Redox processed some Zircaloy cladded fuel, which came from N-Reactor. However, Jungfleisch indicated that the first Zircaloy cladding waste was created in Sept. 1967, and Redox plant shut down in 1966. Other sources (HWN-1991, p. 130) have indicated, on the other hand, that some 269 tons of Zircaloy clad fuel was indeed processed in Redox in 1966. Since the last cladding waste from Redox (CWR) was place in S-107 in 1967q1, we expect that some 18 kgal of CWZr1 sludge would be in the layers of this tank.

The early solids accumulation in Redox waste tanks during 1952-8 is associated with the era where the Redox waste rate underwent substantial change, as noted before. These tanks were also self-concentrating the waste, which increases the tanks' solids load even further. We have used a value of solids volume per cent of 4.4 vol%, which is based on accumulations in SX-105 and SX-111, neither of which tanks were reported to have undergone significant self-concentration over the period in question. These solids per cents are derived based on consistency with the 2.3 vol% that we have found for the second Redox period, R2.

For the second Redox period, solids accumulation in Redox waste dropped to 2.3±1.3 vol%, even as the waste rate for dropped from 2,106 to 1,119 gal/ton from R1 to R2.

Many tanks in S and SX Farms were allowed to self-concentrate and therefore accumulated solids in excess of those from the primary additions. In particular, S-101, S-104, and S-107 were all primary receivers of R1 waste and also were reported self-concentrating waste tanks. Unfortunately, we do not have enough information to always differentiate between the two types of solids accumulation within the waste tanks. However, S-110 was also a primary receiver of R1, but never was reported to have reached boiling. If we assume that the solids for R1 were actually 4.5 vol%, that would provide an estimate for the concentrated solids, RSltCk, in S-101, S-104, and S-107. Thus, we assume that R waste has an implicit component within it that we attribute to the concentrate.

Tank SX-109 accumulated 14 vol% solids from its 1,756 kgal Redox waste. An analysis of the fill history of this tank reveals that it self-concentrated the Redox supernatants, and therefore deposited salt cake. Consequently, we attribute much of the solids accumulation in SX-109 to this salt accumulation and not to Redox sludge. We find that a series of tanks accumulated this Redox salt cake, which amounted to 1,065 kgal in a number of tanks in S and SX Farms. This resulted in a particular waste type, RSltCk.

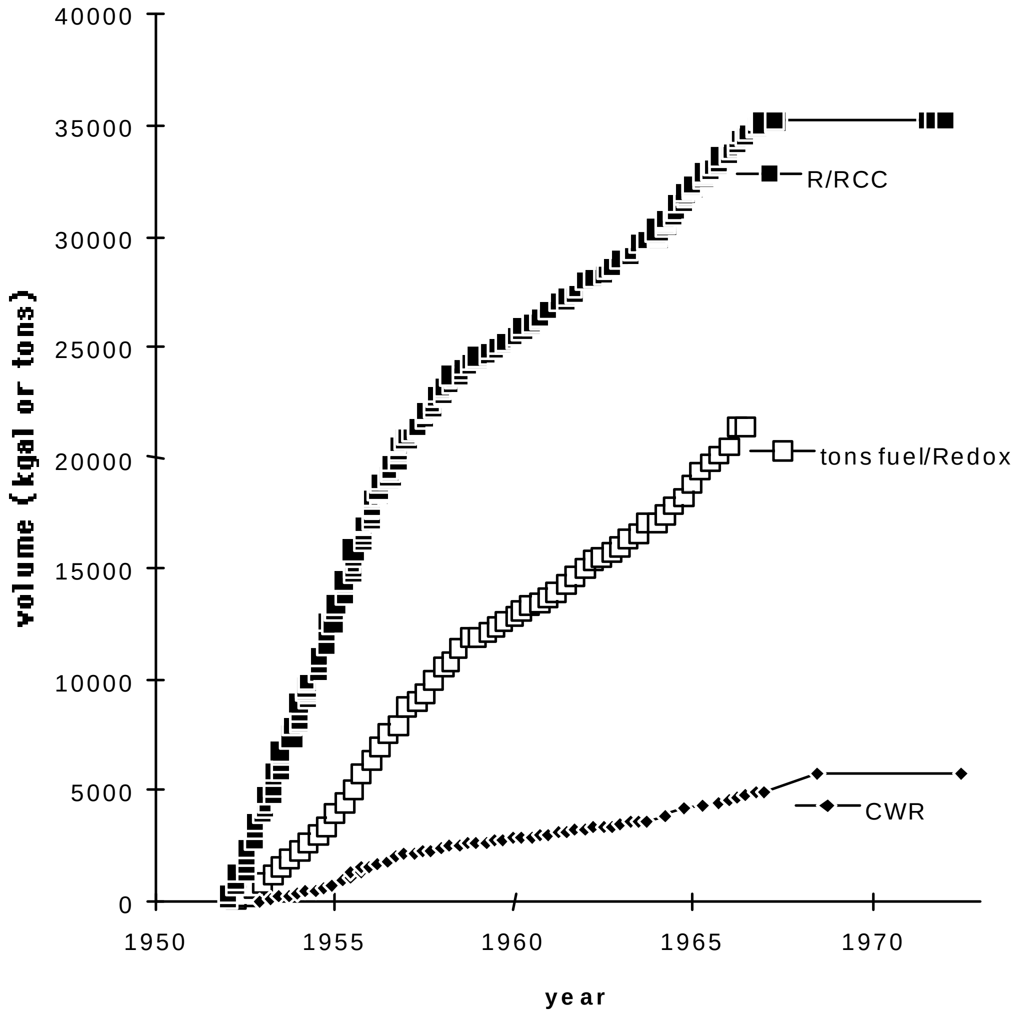

Figure B2. *Total waste volumes for Redox campaign.*

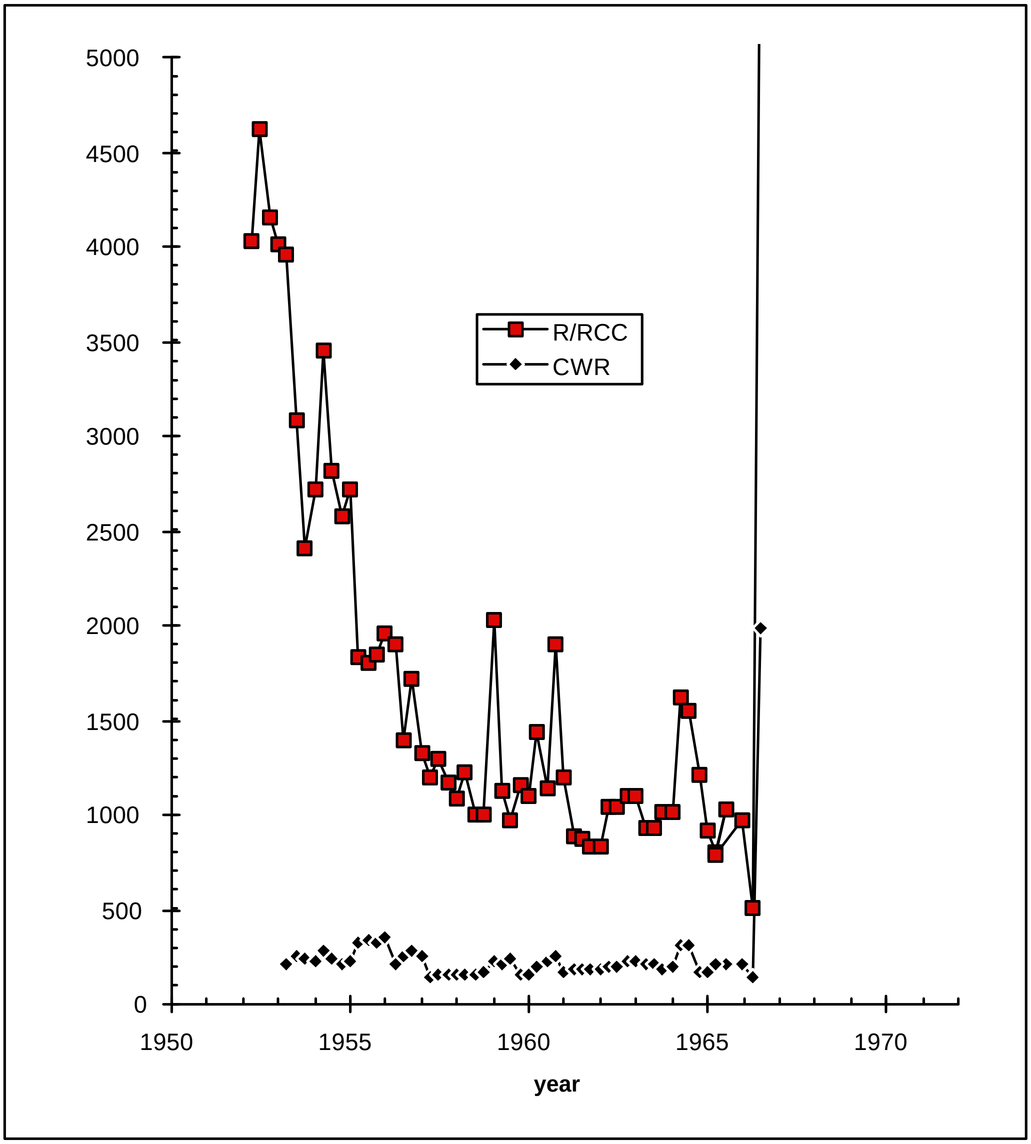

Figure B3. *Waste volume rates for Redox campaign.*

Compositions of Redox wastes were taken from Anderson-91, Jungfleisch-84, Allen-76, as well as published flowsheets.[1] However, there is a difficulty in the amount of silica that is present in the Redox waste tanks is far greater that the amount that is listed as being present in the flowsheet. We have found a similar excessive silica source for Purex and other processes. Thus, we have added an amount of silica to the Redox waste that amounts to 50 mol Si per ton of fuel processed. The fuel that was processed did actually have a silica component, which is listed in the flowsheets as 21 mol Si/ton. At the present time, we cannot explain why the silica is actually much larger.

The amount of iron present in Redox sludge reflects the process vessel corrosion source term that we have found is a significant contribution to the Purex sludges. We have not found any

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[1](a) no author "Redox Technical Manual," HW-18700, July 1951. (b) Crawley, D. T.; Harmon, M. K. "Redox Chemical Flowsheet HW-No.6," October 1960, HW-66203. (c) Isaacson, R. E. "Redox Chemical Flowsheets HW No.7 and HW No.8," RL-SEP-243, January 1965. (d) Jenkins, C. E.; Foster, C. B. "Synopsis of Redox Plant Operations," RHO-CD-505-RD-DEL, July 1978, declassified with deletions.

information about the process vessel corrosion rates during the Redox campaign and have therefore assumed that the rates are identical with Purex.

Appendix C.
Replies to CRS Comments

There are basically two categories of comment within this comment record. Category One comments are those that ask for much more detailed uncertainty estimation for the HLM. Category Two comments are those associated with all other issues. As regards Category One, we have not been directed to expand the workscope for any more detailed uncertainty analyses and so the uncertainties will not be performed for this release of the HLM. Therefore, category One comments will not be fully addressed in the HLM. Category two comments are addressed in the following replies.

A consensus viewpoint of the CRS was that the HLM analysis would be of little value without more detailed uncertainty analyses and the impacts of uncertainty on HLM conclusions. We agree that uncertainty analyses are very important for the HLM and for any model, but such analyses would be beyond the existing scope of the HLM.

We nevertheless believe that there is still value in the existing HLM analysis in the absence of detailed uncertainties. Therefore, on this point we disagree with the CRS. One must recognize that there are multiple unstated assumptions and uncertainties with all existing leak estimates. The HLM is simply an attempt at deriving an independent estimate for a particular set of waste tanks, boiling waste tanks. The HLM analysis was meant to demonstrate the viability of this approach, not necessarily to establish the HLM leak estimates as being definitive.

The following comments are extracts that we believe have captured the primary questions. There was much repetition and overlap among various comments and therefore we have attempted to capture the salient comments in the following extracts. In particular, the need for uncertainty estimates was repeated in many comments and we have distilled and addressed all those comments accordingly.

General Comments from Chemical Reactions Sub-TAP

1) A more rigorous description of the HLM is needed.

Reply:
Agreed. See added detail. We have added the spreadsheet equations and have detailed the important calculations in the text as well.

Comment:
2) Need to consider uncertainties for various input parameters and how those uncertainties affect conclusions.

Reply:
The HLM workscope does not include any extensive inclusion of parametric uncertainties, nor are any sensitivity analyses performed.

Comment:
3) For those tanks that are leakers, what are the uncertainties involved with estimated total leakage volume based on information obtained from monitor wells?

Reply:
This issue is outside the workscope of the HLM. As far as well know, the ranges given in Hanlon for these four tank leak estimates are all based on judgment about soil wetting with waste and not on calculations.

Comment:
4a) With unknown condenser efficiency for reflux, the water and heat losses for that process would be difficult to accurately estimate. Is there really no data available for the reflux conditions?

Reply:
The only data that is available is the amount of condensate in kgal and often this data is incomplete.

Comment:
4b) We consider it incorrect to assume cooling water temperature (e.g. 18 C) and that of the exit air were the same. The statement that, at times tank-specific coolers were required indicates that only 2% of the reflux volume was lost when larger losses could have easily occurred.

Reply:
We agree that these temperatures will in general be different. However, in that absence of data, inclusion of this temperature difference would introduce yet another parameter. Moreover, the amount of conductive heat loss to air is largely already accommodated in the 10 kW to air assumption. Therefore we will continue to neglect this temperature differential and lump the heat loss to air into a single parameter.

Comment:
5) Are all HLM evaporation and reflux rates based on the estimated tank heat load? Are tank cooling rates based on the measured tank temperatures? These bases are not clearly stated.

Reply:
All evaporation and reflux are based on tank heat load as stated in the text, p. 2 paragraph 2, "The HLM then reconciles this volume information with the evaporation rate that is expected based on the total heat load of each tank." There is no tank cooling rate in the HLM per se. There are two heat losses in the HLM: vaporization of water and thermal conduction to ventilated air and ground around the tank. The conductive loss to ground is indeed scaled with tank temperature. The measured tank temperature is used if it is available otherwise it is assumed as stated on p. 4 paragraph 3. We will make this more clear in the text.

Comment:
6a) How do HLM results compare for tanks considered as non leakers?

Reply:
This will be added. In particular, we have evaluated SX-105, which is the only tank with lateral wells that has not leaked, and found that the HLM limit of leak detection is around 0.5 kgal/month, or a factor of three to six lower than any of the other four tanks in this study.

Comment:
6b) We suggest that the HLM should be calibrated by performing calculations (with identical assumptions) for tanks where both the HLM and independent leak volumes can be (or have been derived). Considering the degree of uncertainty that appears to exist in this methodology, these comparisons (with comprehensive consideration of the uncertainties) appear necessary to address the question of whether this methodology can give meaningful results.

Reply:
The HLM is meant to be an independent method for estimating leaks for boiling waste tanks. We compare the HLM to these other estimates but do not calibrate the HLM leak to these other leak estimates. With regards to the amount of uncertainty in the HLM, we recognize that our methodology is very approximate. However, previous estimates are also very approximate. This is why there is a need for independent estimates. The comprehensive handling of uncertainties will require significantly more effort and therefore will need to wait.

Note that the HLM does not show leaks for SX-105, and does not show leaks for five other earlier cooling cycles for tanks SX-111 and SX-112. These other analyses support the interpretation of the HLM unaccounted volume losses as leaks.

Specific Comments from Chemical Reactions Sub-TAP

Comment:
1) Why does the large turnover in tank volumes make determination of tank leaks so difficult?

Reply:
As stated in the text, large amounts of condensate invariably lead to material loss in the condenser system. This material loss will necessitate makeup volumes, and it will be difficult to differentiate this makeup volume from that required to replace material lost to the soil column as a leak.

Comment:
2) What are the uncertainties associated with the “unaccounted” volume losses?

Reply:
We are not going to do much with uncertainties at this stage of the model. However, we know that the smallest leak for the HLM is about 0.5 kgal/mo and this implies at least that amount of uncertainty with the leak estimate.

Comment:
3) How do results derived for other SX Farm tanks compare with those of the four tanks in the HLM?

Reply:
We have added one additional nonleaking tank (SX-105) to the HLM analysis for comparison. The remaining comparisons will require additional effort and therefore workscope. Generally, there are unaccounted losses in non-leaking tanks, and these losses define the limits of the HLM.

Comment:
4) How are sludge radionuclides such as Sr-90 partitioned compared to supernatant radionuclides such as Cs-137?

Reply:
When process waste is placed into a tank, both soluble and insoluble radionuclides are added together. Normally, the waste remains in each tank for at least a year to allow the short-lived radionuclides to decay. Thereafter, for supernatant transfers the HLM simply partitions the remaining heat equally between the supernatant and sludge.

Comment:
5) How does partitioning between sludge and supernatant affect the HLM? What uncertainties does the HLM partitioning assumptions introduce?

Reply:
Sludge/supernatant partitioning is only important for waste transfers out of the tank and therefore is most important in the tail of each tank’s cooling curve. This will be clarified more in text.

Comment:
6) How was evaporation modeled for this analysis?

Reply:
Evaporation was modeled very simply in the HLM. All heat above the conduction losses was removed by vaporization of water at 100 C.

Comment:
7) What is the uncertainty associated with estimating evaporative losses?

Reply:
Once again, the HLM analysis was not scoped to perform detailed uncertainties. However, we feel that the primary uncertainty in the evaporative loss is with our assumptions about heat load and conductive loss in the first place. Then, the heat of vaporization of water from waste is different from that we assumed, the temperature of the dome vapor will vary depending on many factors such as ambient air temperature, season of the year, time of the day, and ventilation rate. And then there are the losses of condensate from the system. These losses need to be made up with water additions.

Comment:
8) What fraction of “unaccounted volume increases” could be ascribed to uncertainties in information and methodology? What is “normal volume loss” and what are “inaccuracies of the HLM?” What are the criteria for deciding between “inaccuracies of the HLM” and “waste inventory that has been lost to the soil through a breach in the tank liner”?

Reply:
Uncertainties are beyond HLM scope. Normal loss is assumed to be that associated with makeup volumes and is not an HLM parameter at all as these losses are made up by water additions. There are no criteria for deciding between HLM inaccuracies and leak inventory. Leak inventory is defined by the leak parameters, which are in turn adjusted to eliminate unaccounted volume losses over some period.

Comment:
9a) What are the uncertainties associated with the (calculated?) “evaporation rate” and unaccounted volume loss”?

Reply:
Uncertainties have not been estimated. The evaporation rate is indeed calculated.

Comment:
9b) How would “a significant temperature decrease” cause “more evaporative heat loss”?

Reply:
A reported tank waste temperature decrease does not cause evaporative loss, it simply indicates it. That is, evaporative cooling will lower the waste temperature.

Comment:
10a) It is very difficult to keep track of which values are measured, which are assumed, the basis for the assumptions, and the uncertainties associated with the assumptions and measured data.

Reply:
We will attempt to clarify this more in the methodology section. We do not have uncertainties for the data nor for our assumptions more than what is already stated in the report.

Comment:
10b) Are evaporation rate and unaccounted volume loss measured or estimated by HLM?

Reply:
Evaporation rate is calculated based on heat load. Unaccounted volume loss is calculated based on reported tank volume, volume additions and removals, and evaporation. We will try to make this clearer in the methodology.

Comment:
11a) It is stated that tanks that were cooled by evaporation are assumed to have been on a reflux condenser system. How can this assumption be confirmed?

Reply:
Since there are reports of condensate removal and water additions for these tanks, this is the basis for these assumptions. Presumably, there are logbooks associated with these operations that we have not yet found.

Comment:
11b) Can details of the process be clarified? Is it possible to determine which tanks were on separate individual coolers and which were on combined ventilations systems? How will these uncertainties impact the results of the HLM?

Reply:
We have some more detailed information as stated in the text. That is all we have at this time.

Comment:
12) What is there to indicate that leak rates derived from HLM are meaningful?

Reply:
The scope of the HLM was to provide an independent estimate of leak volumes for these aging waste tanks. The finding of much larger than anticipated ground contamination around SX-108 certainly suggests that the previous leak estimates may have been too small. This question of how meaningful is the HLM is of course relevant to all leak estimates. How do we know any of these estimates are meaningful? In the end, both end up with a contaminated soil column, which we already know from various drywell observations.

Comment:
13) Is it possible to determine whether the condensate in SX-106 was used for makeup additions? Won't this uncertainty affect the results given by the HLM? What is the basis for a 2% loss and how does it affect the HLM leak estimates?

Reply:
There are reported transactions from SX-106 to each of these tanks. Comments in Anderson list SX-106 as a condensate receiver. The 2% loss for makeup was used to illustrate a point and does not impact the HLM.

Comment:
14) Are several "periods of low reflux" used for determination? If not, extrapolation would appear to be uncertain. If more than one "period of low reflux" is used, what is the resulting standard deviation considering each period as resulting in one value?

Reply:
Each period of low reflux has volumes reported every quarter. Therefore, each quarter's volume measurement contributes to the accumulated volume discrepancy. However, the overall driver in the HLM is the loss expected based on calculated evaporation based on heat load. Note that each of these tanks have already been assigned as leaking and therefore the HLM did not invent these leaks. The HLM simply interprets unaccounted volume losses and extrapolates them over time.

Comment:
15) What uncertainty is associated with the HLM assumptions of 20 kW conductive heat loss to ground and air?

Reply:

The traditional understanding of SST's at Hanford has been that their passive heat loads were always limited to 40,000 Btu/hr or 10 kW. This kept waste in that tank below boiling (100 C) for the entire year. Ventilated DST's are allowed twice that heatload at 20 kW for a similar reason. Therefore, the HLM simply uses these values as heat loss assumptions. These assumptions are most important in the tails of the tank waste cooling curves and this is of course the period for which the HLM is calibrated. Note that there are times when the waste cooling curve matches that calculated quite well. Otherwise, we have not evaluated this uncertainty of this nor its impact on the HLM leak estimates.

Comment:
16) We suggest a better description of assumed tank failure modes. Figures illustrating the scenarios considered would be helpful.

Reply:
The tank failure modes are not important for the HLM. We placed this in the report to provide some background.

Comment:
17) We suggest more information about the potential plume geometry.

Reply:
Discussion of plume effects was only meant to be qualitative and does not impact the HLM. Therefore, more discussion is not appropriate. There are other more appropriate references for plume geometry.

Comment:
18) The uncertainties associated with each assumption must be given if the validity of the HLM methodology is to be established.

Reply:
We agree that uncertainties are very important to completely validate HLM results. However, uncertainties are beyond the existing work scope. We nevertheless feel that there is still value to the HLM report despite these deficiencies.

Comment:
19) How were modes of failure and bottom bulges observed? What were the level changes associated with bottom bulges (and their disappearances) for SX Farm tanks?

Reply:
Bottom bulges were reported with sludge height measurement differences among risers. Also, bent thermocouple trees and other equipment were associated with bottom deformations. No liquid level changes have, to our knowledge, ever been attributed to bottom bulges. However, the volume changes that occurred for SX-108 in the 1959-61 period coincide with the reported bulge that then relaxed back into place.

Comment:
20) What is the depth of the drywells? Do any drywells extend horizontally beneath the tanks? A schematic figure showing the relation between waste level, assumed waste level, drywell locations, assumed plume boundary, assumed soil porosity, soil permeability, etc., would be extremely useful.

Reply:
Drywells range in depth from 35 to several hundred feet. We will add references to reports that detail these issues since none of these issues impacts the results of the HLM. There are drywells underneath tanks SX-107 through SX-115 that are termed laterals. They extend from caissons placed among the tanks with three laterals underneath each tank.

Comment:
21) We consider a better description for the reflux system and associated model assumption to be required. Since a large fraction of the HLM uncertainty may be associated with reflux systems, we suggest attempts be made to identify better information concerning the performance of the reflux systems.

It is assumed that the dew point of the condenser is 65 F. This requires either a refrigerated system or a very high performance cooler...Accurate knowledge of the ventilation system would appear to be critical...Do records for any of the tanks indicate local steam venting?

Reply:
We agree that better information would be desirable. However, we only have limited information and are necessarily limited by that until better information is available. More precise details of the reflux system may or may not affect the HLM results. The largest fraction of uncertainty is most likely associated with the tank heat load and conductive loss and not on condenser efficiency.

Comment:
22) What is the evidence supporting description of a leak by the two parameters, leak size and elevation? Why would leak rate be expected to be constant for any significant period of time for a given driving pressure?

Reply:
These leak parameters describe the simplest possible leak: a leak size that scales with hydraulic head and a leak elevation that defines the height of the leak within the tank. These parameters are used because they are the simplest. To start and stop a leak more than once requires yet another set of assumptions and it is not clear how we would assign those additional parameters except as we have for SX-112. In this case, a cooling period in between the two leak periods strongly suggest a two part leak.

Comment:
23) Paragraph 4 p. 5 seems to be contradictory. [This paragraph talks about self-sealing leaks.]

Reply:
Corrected.

Comment:
24a) Why is the tank waste temperature assumed? Is it not known?

Reply:
We have limited tank waste temperature data. When it was available, we used it. Otherwise, it was assumed.

24b) What is meant by chilled air that could either be vented or returned to each tank? Was this a closed system?

Reply:
Ventilation systems were (and still are) operated in a partial recycle mode. The amount of return was often adjusted according to some particular criteria the details of which we have not found. For example, Tank C-106 is operated today with a chiller where around 60% of the vent volume is returned to the tank and only 40% is replaced by incoming air.

Comment:
25) Last paragraph p. 5 is not clear or helpful.

Reply:
This paragraph was meant to describe the gaps in the tank level data do not allow us to quantitatively ascribe level variations to waste evaporation. We will try to make is clearer by comparing periods where monthly instead of quarterly data were reported.

Comment:
26) The section describing the reflux model does not appear to support use of HLM to quantitatively describe a leak rate or volume.

Reply:
Once again, we feel that the heat load and conductive loss of the tank are the most important sources of uncertainty. There are many deficiencies in the HLM reflux model as pointed out in this section. We still feel that the tank heat load is likely the most important source of uncertainty.

Comment:
27) see 18), where need for uncertainties is reiterated.

Comment:
28) Why aren't unaccounted volume gains considered due to accounting uncertainties? Why aren't unaccounted volume losses also due to accounting uncertainties?

Reply:
Unaccounted volume gains are assigned to water additions by the HLM. Unaccounted losses above those expected from evaporation could correspondingly assigned to waste losses by transactions. The HLM is simply attempting to bound those unaccounted losses by reconciling evaporative loss with reported tank volume.

Comment:
29) Condensers are well known for having tubesheet failures. What is the basis for the assumption that a makeup of 2% of the reflux rate was appropriate or conservative. It would not take a very large leak in a condenser to add a significant volume of water to the tanks. Are there maintenance records relevant to this potential behavior available?

Reply:
The HLM does not depend on the 2% loss that is stated and this is only used for descriptive purposes. There are no records yet available for SX Farm operation with this kind of detail. However, 242-T and 242-S in-farm evaporators reported a 5% materials accountability criterion for their operation. Because the in-tank evaporation for SX Farm was a much more complex than the in-farm evaporators, this suggests that SX Farm condenser operation probably used a materials balance on the order of 5-10%.

Comment:
30) What comprises a bulge? How are they observed? Do bulges always have associated leaks? A schematic would be helpful.

Reply:
Bulges were associated with sudden changes in sludge weight measurements for particular risers. Therefore, their detection was dependent on the performance of a sludge height measurement performed close to the deformation. They also causes equipment in risers to become bent or deformed and all reported tank bottom bulges (SX-108, SX-113, A-105) have also been associated with leaks. It is therefore likely that there were other tank bottom bulges and that those bulges were never actually detected.

Comment:
31) Internal tank bottom deformations would have caused level changes. Did the HLM methodology include estimates for these factors? Could a relaxing bulge a the end of a high temperature period have been interpreted as a leak?

Reply:
Undoubtedly the tank bulges resulted in waste level changes. For example, the level changes for SX-108 in 1959-61 may have been at least partly due to a bottom bulge. A relaxing bulge could very well have been interpreted as a leak. However, the HLM does not include any compensation for level changes due to bottom bulges.

Comment:
32) What is the relation between lateral well activity and tank level stability (or leak rate) as a function of time?

Reply:
Within the HLM, there is no quantitative link between lateral well activity and tank leak rate. There is only the qualitative association that continued increase in lateral activity is associated with periods of time that each tank leaked waste by the HLM.

Comment:
33) What is the basis for the assumption that each leak rate by be ~50% lower than that stated within the HLM.

Reply:
This assumption was an attempt to get at the uncertainty in the tank heat load as well as the uncertainty in the leak “duty factor”, or the period of time that a tank may actually have been leaking. This is not a quantitative uncertainty and is only a judgment at this point. Any quantitative description of uncertainty will require more effort and therefore expanded work scope.

Comment:
34) Notes on each tank do not appear to support the use of HLM for describing leak rate or volume.

Reply:
This information was meant to provide some background. The timing of the ground contamination and bottom bulges is important for the HLM, but otherwise the timing and leak rate are independent of past estimates.

Comment:
35) The HLM leaks are calibrated by reducing unaccounted volume losses by 80%. Why 80%?

Reply:
There is a variation of these unaccounted losses and therefore we did not feel justified in pushing it to 100%. An 80% value seemed reasonable in light of the other uncertainties present within the model. The filled diamonds on each plot show the remaining unaccounted volume losses for each tank (see comment 41).

Comment:
36) There does not appear to be a dependable basis for the leak rate and start and end times. Need to specify the assumptions and their uncertainties.

Reply:
The assumptions will be stated. As mentioned previously, uncertainties would require more work and are out of the scope of this effort.

Comment:

37) Factors that should be used to correct for soluble radionuclides are not clear.

Reply:
We will try to make them clearer.

Comment:
38) The description of the Redox process needs more detail. Need to state the bases for the earlier report and their uncertainties.

Reply:
We will add more detail about the HDW model bases. The uncertainties will have to wait.

Comment:
39) What is the basis for adding the amount of silica of 50 mol per ton of fuel processed?

Reply:
This is described in the HDW report and is related to that fact that silica shows up in waste analyses but was never added during processing. The primary known source of silica is from the bonding agent that was used in sintering fuel pellets. This addition is meant to reflect other silica sources that were unaccounted.

Comment:
40) We suggest a more complete description of how the spreadsheet was calculated.

Reply:
We will include these details.

Comment:
41) Figures require additional explanation. What is the significance of the filled diamonds? Why don’t figures seem to illustrate the HLM results?

Reply:
Figures do show HLM results as the greyed line. This is the calculated tank volume in the absence of a leak. The filled diamonds were meant to represent unaccounted volumes that remain since only ~80% of the unaccounted volumes were assigned to each leak.